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Kinetics of Materials at Extreme Conditions: Understanding the Time Dependent Approach to Equilibrium at MaRIE

Unclassified

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Section 1

Kinetic Response of Materials at Extreme Conditions

1.1 Overview

The National Nuclear Security Agency has recently recognized that a long-term need exists to establish a stronger scientific basis for the assessment and qualification of materials and manufacturing processes for the nuclear stockpile and other national security applications. These materials may have undergone substantial changes with age, or may represent new materials that are being introduced because of difficulties associated with reusing or recreating materials used in original stockpile components. Also, with advancements in manufacturing methods, the NNSA anticipates opportunities for an enhanced range of control over fabricated components, an enhanced pace of materials development, and enhanced functionality. The development of qualification standards for these new materials will require the ability to understand and control material characteristics that affect both mechanical and dynamic performance. A unique aspect for NNSA is that the performance requirements for materials are often set by system hydrodynamics, and these materials must perform in extreme environments and loading conditions. Thus, the scientific motivation is to understand "Matter-Radiation Interactions in Extremes (MaRIE)."

Given this need, it was natural and an opportune time to hold a workshop to consider the experimental requirements for developing a scientific understanding of the time dependent response of materials. Like most natural phenomena from earthquakes to cancer, systems under dynamic loading conditions evolve in a complex, time-dependent fashion. Equilibrium and continuum based models are not adequate for understanding and predicting these phenomena. The immediate, short time scale response that initiates over nanometers eventually leads to correlated effects that occur through the formation, growth, and evolution of material interfaces like grains and cells at the micron to millimeter distances. The study of the timescales at which these phenomena proceed and evolve towards equilibrium is known as "kinetics."

This report describes the thinking and conclusions that emerged during the workshop. The scientific opportunities are posed as grand challenges within the following topical areas:

- Phase transitions between solid, liquid, and plasma states of matter in dynamic, non-equilibrium environments
- Mesoscale interactions at length scales that bridge atomistic and bulk continuum response common in foams, granular systems, or architectured materials
- Chemical kinetics under extreme conditions
- Plasticity and fracture mechanisms that dissipate energy and evolve material interfaces dynamically

Within these fields of material science, the change in state of a material is transmitted through an interface that separates the initial, intermediate, and final states. In order for our community to understand the time dependent evolution of a material, we need to know how these interfaces form, interact with each other, and sweep out volume as a function of time.

- *Crosscutting Theme: Understanding the formation, interaction, and evolution of material interfaces during dynamic processes.*

Beyond the physical process of how a transition between material states occurs, a fundamental aspect of a process that occurs out of equilibrium is that entropy is created and energy is dissipated. We need to understand the dominant dissipative mechanisms associated with the non-equilibrium nature of the process in order to fully understand the driving forces and limitations for high-rate processes.

- *Crosscutting Theme: Determining the dominant dissipative mechanisms associated with non-equilibrium dynamic processes.*

These two cross-cutting themes have in some sense been at the center of developments in kinetics since metallurgists began thinking about the grain sizes in steels and are now being investigated in greater detail for problems like shock initiation in high explosives, Figure 1.

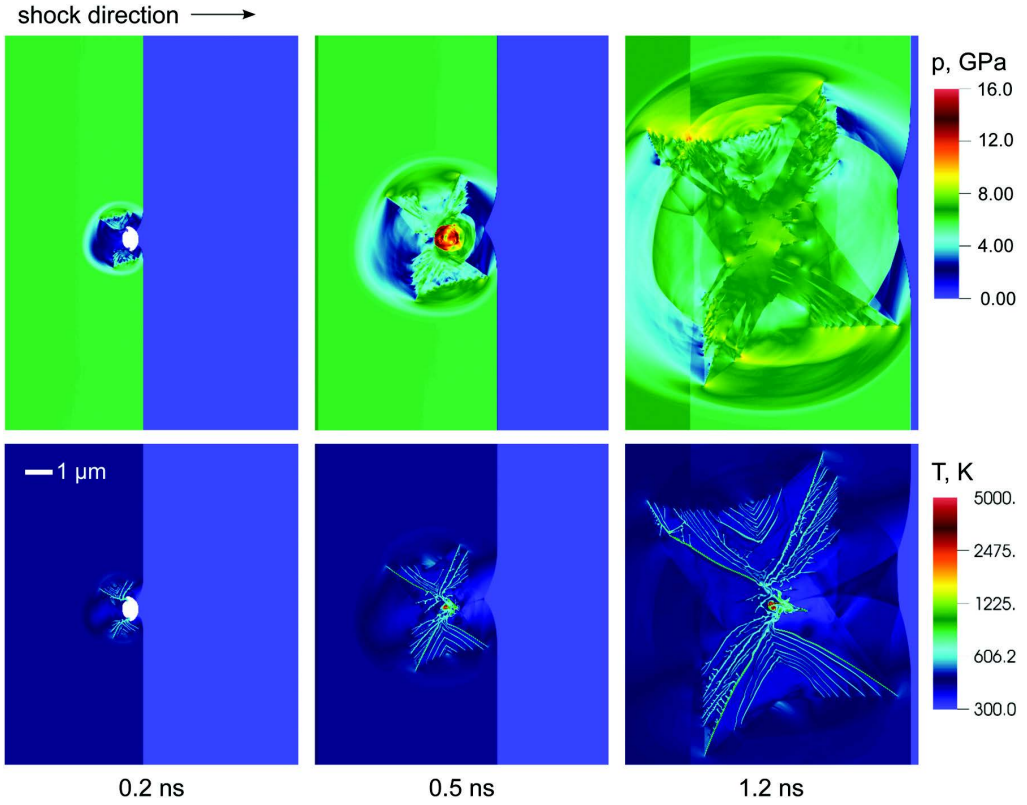

Figure 1.1: Simulated pressure and temperature fields that are generated when a ~10 GPa shock wave collapses a single pore in a HMX crystal. The time origin coincides with the arrival of the shock wave at the left hand side of the pore. This simulation shows have mesoscopic features can lead to much larger and longer timescale effects such as detonation. Figure from Austin et al. JAP **117**, 185902, 2015

However, there is a consensus within the communities that we need major technological advances to put us over the edge, in terms of our ability to answer our grand challenge science questions. Without immediate regard for cost or technical capability, we propose that three major advances in experimental techniques are necessary to address all our science questions:

1. Three-dimensional microscopy and tomography to directly image interface formation and motion.
2. To understand the dominant dissipative processes associated with rapid dynamic processes, we will need temporally and spatially resolved temperature measurements (or entropy, but temperature is commonly considered easier to measure).
3. To resolve the physical processes that evolve over disparate timescales, we will need a suite of pressure drivers that can apply large stresses (from 1 GPa to 500 GPa) over timescales of nanoseconds to microseconds.

In the rest of this document we describe the grand challenges questions within each field, the current technical limitations causing us not to be able to address the grand challenge questions, first experiments using the desired new technical capabilities, and the specific experimental needs for understanding kinetics of materials in the following topical areas,

- Time Dependence of Phase Transitions
- Chemical Kinetics
- Rate-Dependent Plasticity
- Waves in Mesoscale Structures and Heterogeneous Media

Although these topical areas are generally quite distinct in theoretical basis, one will be able to trace the underpinnings of the two crosscutting themes throughout the following four sections.

Section 2

Time Dependence of Structural Phase Transitions

By Carl Greeff and Jon Eggert

2.1 Introduction

As the intrinsic state variables evolve, materials transform between disparate crystal structures towards the one with the lowest Gibbs free energy. The crystal structure has important implications for both the compressibility and the shear strength, both critical aspects of determining the high rate hydrodynamic response of materials at extreme conditions. As the transformation between crystal structures occur by correlated motion of the atoms, there is a finite timescale associated with the transition, which for high-rate flows can be an appreciative fraction of the hydrodynamic timescale. Because of the importance of crystal structure and the potential time-dependence associated with reaching the equilibrium phase, our community is intensively investigating the kinetics of structural phase transitions.

Traditionally, dynamic compression experiments on materials with phase changes have been interpreted using macro-scale models. These treat phase fractions as continuum variables that evolve towards equilibrium under phenomenological rate laws. The rate expressions have parameters that must be adjusted based typically on wave profile data. This modeling level allows some information to be inferred about the processes within the sample based on surface velocity measurements. However, such models contain no information about microstructural variables such as second phase domain size. Their parameters are not predictable based on other measurements or first principles theory. No prediction is possible for the effect of impurities or changes in the initial microstructure. The coupling of shear deformation to solid-solid phase transitions is known to be important, but it is difficult to incorporate into a model at this level.

What are needed are physical models that have the correct internal state variables, and whose parameters have well-defined meanings, so that they can be estimated. We also require that the phase transition model provide informative state variables to other material models, such as strength, ejecta, and damage.

There are efforts currently underway to improve the physical foundation for model of dynamic phase change. For instance, J. Belof described an approach based on classical nucleation and growth theory aimed primarily at dynamic solidification, see Figure 2.1. The principal objects in this theory are the energetics and mobilities of interfaces between solid domains and the surrounding liquid. Solid-solid phase transitions involve the same issues, but bring in additional complications, including long-range strain fields and their associated energies, which tend to

inhibit complete transformation, and play an important role in hysteresis. In addition, interface energies and mobilities are expected to be anisotropic.

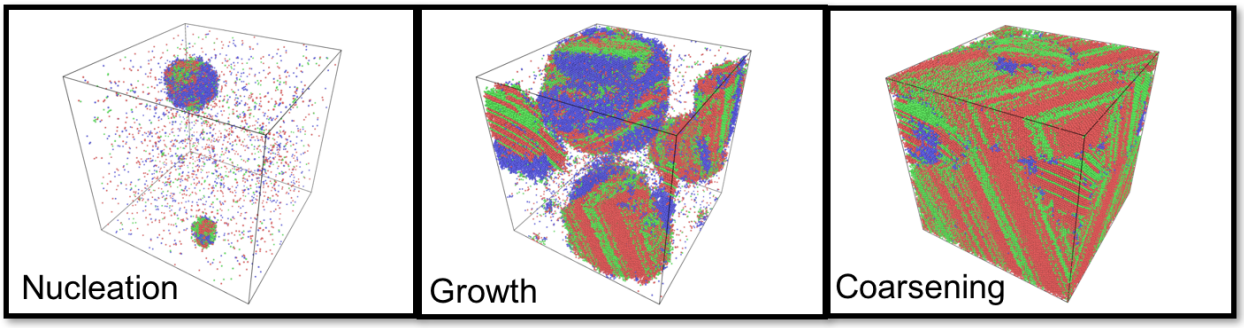

Figure 2.1: MD simulation of the nucleation of solid grains, growth and interaction of the solid grains, and coarsening of the phases into the final microstructure for the dynamic solidification of copper. This simulation shows how the microscopic atomic motion at growing interfaces can have significant influence on the final crystalline microstructure. Figure from Luis Zepeda-Ruiz (LLNL).

Models have been constructed which attempt to deal with these complexities, however, confronting them all at once is probably impossible. What is required is experimental guidance as to what are the important meso-scale structures and processes.

Focused dynamic compression experiments offer the opportunity to define the onset of pressure-driven non-equilibrium thermodynamic states leading to structural phase transitions. The standard model for phase transition kinetics include nucleation, growth, and coarsening processes, which are all dominated by different interfacial energies, interactions, processes. Unfortunately, traditional experimental probes are largely constrained to integral measurements like surface motion, which have a difficult time constraining theoretical modeling.

Free electron lasers (FELs) offer the opportunity to probe these structural transitions on an atomic scale with unprecedented accuracy in a time and spatially resolved manner. We are just now beginning experiments with the potential to inform theoretical kinetic modeling of phase transitions at new science FEL and synchrotron facilities such as LCLS, DCS, EXFEL, and the ESRF. We believe that MaRIE will be the next evolutionary facility that will draw upon the ongoing experiments at these facilities while opening the potential for revolutionary comparison over a very wide range of driver and source capabilities. Our overarching goal is to interpret and incorporate the experimental data into a general, accurate, rate-dependent, and preferably predictive infrastructure that can be incorporated into hydrocode simulations.

2.2 Grand Challenges:

Grand Challenge 1: *Determine the rate-limiting processes during all stages (nucleation, growth, and ripening) of pressure-driven melting, freezing, and solid-solid phase transitions.*

There are a number of fundamental questions that can only be answered experimentally about how materials progress toward equilibrium following a phase transition.

- What are the independent variables affecting the rate of phase transitions?

These may include the nature of nucleation (homogeneous vs heterogeneous), interplay of deviatoric stresses and strains, energy flow associated with interfacial motion, amorphous and metastable phases, anisotropy in crystalline nuclei and phases, and the role of impurities.

- What is the microscopic state of the material throughout a phase transition?

The microstructure (grain size, preferred orientation, dislocation density, etc.) of the new phase will greatly influence the bulk material properties and response of the resultant material.
It is important to understand the evolution of the microstructure in order to develop complete models for the response of driven materials. These studies require a broad range of drivers with similar diagnostics in order to span the very wide range of time scales for important processes.

Grand Challenge 2: *Design and recover new materials by harnessing phase transition kinetics under dynamic compression.*
Since the discovery of synthetic diamond, it has been a goal in high-pressure science to synthesize other useful materials that can be recovered to ambient conditions. Data obtained from MaRIE should prove invaluable in achieving this goal.

- How does the final material state depend on compression and de-compression rates and profiles?

It may be that induced stress states, impurity distributions, or novel release paths can be usefully employed to extend the metastability or recoverability of novel phases.
In situ studies with spatial and temporal resolution obtained throughout the compression and release will offer invaluable guidance in our quest to produce new materials.

2.3 Approach and Impact

The most common experimental configuration will be x-ray diffraction and imaging of a planar pressure drive on a planar tamped sample. We require a broad range of pressure drivers including laser, pulsed power, gas gun, high explosive, and dynamic diamond-anvil-cell to cover a very wide range of strain rates from 10^8 to 10^2 s^{-1}. A tunable pressure drive history will be required for all experiments as a combination of shock and ramp compressions is the simplest way to control the P-T thermodynamics of the final state. The pressure drive needs to be highly accurate and repeatable in order to demonstrate reproducibility.

The attributes of a high-energy FEL are ideal for these studies. High temporal precision will allow detailed information about the transition pathways. High photon energy is critical for several reasons: to increase the transmission of x-rays through thick high-Z samples, to extend the accessible Q-range for diffraction giving dramatically improved amorphous, PDF, and crystalline diffraction data quality, and to reduce the intrinsic background level from high energy drive such as laser and pulsed-power drivers by aggressive filtering at the detectors. We would also very much like to do xray spectroscopy (XRS , XANES, EXAFS, XRF, and possibly XR Raman) to improve the determination of short range order, temperature, and chemical state of the material phase and impurities.

2.4 Current Limitations

Current facilities for this type of experiments have a variety of limitations. Traditional drivers are limited in probing crystalline structure by the available x-ray sources: metallic fluorescence lines for lasers and pulsed power, spark gaps for gas guns, and temporal resolution for DACs.

Emerging x-ray FEL and synchrotron sources are limited by the quality of the pressure drivers. The MEC hutch at LCLS has a laser with limited energy and pulse shaping capability. The DCS beamline at APS has a state of the art 100J laser, an excellent gas-gun facility, and a 15 g HE chamber. But is limited by the bandwidth required for sufficient photon numbers for single bunch diffraction, maximum monochromatic photon energy, and by the driver capacities (energy and rep. rate, bore-size, and mass for the laser, gun, and HE drives respectively). The HED instrument at the EXFEL will soon have a 100J IR laser operating at 10 Hz, but will be limited by maximum laser and x-ray photon energies.

In general, current x-ray sources have complementary limitations: FELs have high coherence and narrow bandwidth, but cannot incorporate a wider bandwidth for absorption spectroscopy, while synchrotrons have a broader monochromatic bandwidth for single-bunch diffraction limiting diffraction peak shape analysis and complex structural analysis, but have been shown to produce excellent single-bunch absorption spectroscopy. All emerging facilities have the limitation of forbidding work on radioactive materials.

2.5 First Experiments:

2.5.1 Measure the microstructure and phase fraction through a solidification phase transition as a function of time and strain rate.

This experiment will be performed by shock melting into the liquid phase, ramp compressing into the solid phase. Complete melting and the liquid structure and density will be determined by liquid diffraction. Upon compression into the solid stability regime the phase and phase fraction will be determined by the transition from liquid to solid diffraction and the crystalline microstructure will be probed for grain size, preferred orientation, and deviatoric strain by peak shape analysis.

Similar experiments will be repeated on all pressure drivers to explore the behavior over a wide range of strain rates. In order to perform the cross-platform comparisons desired, each platform needs to have excellent pressure-drive shaping, reproducibility and control at the $\Delta P/P<2\%$ level.

2.5.2 Develop ramp compression and quenching to recover metastable phases.

This experiment will explore the hysteresis and recoverability of new phases by designing compression and release pathways to expand the stability range of the new phase. This is a completely new type of research enabled by emerging technologies and it is very difficult to predict requirements beyond those for the preceding experiment.

2.5.3 Multi x-ray beam and/or electron beam scattering for grain orientation microstructure studies.

We would like to perform tomography using crossed x-ray or x-ray / electron beam tomography in order to resolve the spatial distribution of an emerging phase.

2.5.4 Track the motion of impurities during a dynamic phase transition using x-ray spectroscopy.

By employing species-specific spectroscopy we will explore the distribution and short-range environment of impurities following a phase transition.

2.6 Requirements (exceptional beyond baseline)

- 70 keV photons to reduce heating in the sample for multi-pulse experiments.
- Pressure-drive shaping, reproducibility and control at the $\Delta P/P<2\%$ level to reach desired thermomechanical states

Section 3

Rate Dependent Plasticity

By Ellen Cerrata and James Stolken

3.1 Introduction

Under any loading condition, solid materials dissipate energy through plastic and fracture processes. These processes evolve the strength and damage properties of the material. In the extreme environment of dynamic loading, these processes are not well understood and therefor dynamic strength and damage is poorly predicted. A robust predictive understanding of this evolution could enable the design of next generation damage tolerant materials, accelerated certification of new materials, and enhanced confidence in material performance. This capability has not been realized because we lack characterization tools to explore stress state and structure under extreme conditions.

Here an approach for development of a predictive understanding of material strength and damage under extremes of dynamic loading is discussed. Limitations in current experimental and theoretical tools for such work are described. First experiments and definitions for the required diagnostics are proposed to address long standing questions associated with material response far from equilibrium. Through this discussion, a consensus of experimental drive platforms and multi-probe diagnostics necessary to map structure/property relationships is presented.

3.2 Grand Challenge

Grand Challenge: *Develop a predictive understanding of material strength and damage at high strain-rates ($>10^4$ s^{-1}) and pressures (>1 GPa).*

3.3 Approach

Map material structure to property relationships by spatially and temporally resolving the evolution of microstructure, strain-state, and temperature over relevant length (<1 μm) and time (<1 μs) scales in three dimensions.

3.4 Impact

Predictive understanding of material strength and damage response under extreme conditions of dynamic loading enables:

1. Identifying links between materials processing routes and failure modes to design the next generation of damage tolerant materials.
2. Assessing the role of material structure on performance to accelerate certification
3. Increasing confidence in material performance margins by reducing uncertainties.

3.5 Background

Understanding material strength and damage at high pressures and strain-rates is complicated by the explicit stress-state and load-path dependence of their underlying phenomenology. The 6-dimensional nature of stress and the multiplicity of mechanisms such as dislocation glide, twinning, phase transformations, etc, through which it can affect the permanent deformation associated with material strength represented fundamental challenges to understanding the mechanics of strength from the 1920's through the 1950's. These challenges were largely met and overcome through careful experimentation that controlled the stress-state coupled with material characterization to identify the deformation mechanisms under static or simple dynamic loading followed by ex-situ characterization. The dynamic and transitory nature of high-pressure and high-strain rate material states drives the need for in-situ material characterization and complicates full-exploration of the multidimensional stress dependence of material strength under these conditions.

Material processes during deformation may result in the evolution of dislocation structures, material temperature, twins, recrystallization, void nucleation and growth, shear localization, cracking of inclusions and grain boundaries; ultimately leading to material failure. These processes depend upon the deformation path and subsequent microstructural evolution resulting in material failure being, in general, dependent upon the entire deformation history. It is these considerations that necessitates spatially and temporally resolving the evolution of microstructure, strain-state, and temperature over the deformation path. The size of relevant microstructural features set the length scale (<1 μm) and the loading rates of interest set the time scale (<1 μs). Coupled with the three-dimensional nature of the loading and the microstructure motivates the proposed approach to the above Grand Challenge.

3.6 Current Limitations

Currently, the study of evolving plasticity, particularly under the extreme environment of dynamic loading, is limited by the temporal and spatial resolution of existing diagnostic probes. Where certain probes do have the temporal or spatial resolution, the availability of 3D data can also be limited because angled or split probes are not available. In general, existing probes cannot be *tailored* in terms of energy, timing, or exposure. The current facilities for such studies are in high demand and access is limited in terms of both time and materials handling which leads to a lack of systematic studies to firmly anchor data within parametric space. These same facilities often do not lend themselves to a robust set of drive conditions and we broadly lack the analysis tools to efficiently correct and interpret the data collected from such experiments.

3.7 First Experiments

3.7.1 Planar Loading

To address long-standing questions regarding the structure of a material at pressure and how that structure deterministically links to observed strength and damage of the material, we propose to perform relatively simple planar loading experiments. These experiments should be agile in nature and therefore incorporate the ability to access moderate to extreme conditions of temperature and pressure to allow for full definition of the structure-property parameter space. Uni-axial strain experiments as well as those that incorporate significant shear loading should be possible to allow for a step-wise, fundamental understanding of the effects of integrated loading conditions. To obtain the necessary understanding required to link microstructure to strength and failure, the stress-state, dislocation density, temperature, crystal structure, crystallographic texture, 3D microstructural evolution, early stage void and/or crack growth, and late stage damage kinetics should be measurable with sufficient temporal and spatial resolution. The details of these measurements are discussed in the diagnostics section of the experiment description.

Drives:

A robust set of loading conditions for this type of experiment can be achieved with a suite of traditional platforms, including: laser, direct high explosive, gas gun (single and two-stage), as well as electro-magnetic drives. To facilitate addressing the fundamental and applied science questions over the pressure–temperature parameter space of interest, peak stresses achievable with these platforms should range from 1 to 500GPa. Pulse durations should range from 1ns to 1μs. Additionally, specimen assemblies or targets should be designed to maintain the high-pressure state for extended time durations. To provide enhanced shear loading over traditional plate impact type experiments, the capability to design and build graded density flyer plates is needed for use in guns with keyed barrels.

Diagnostics:

For the measurements listed above, the following diagnostic probes are suggested to be simultaneously available. It is envisioned that experiments using all or only a subset of these probes will be proposed.

Table 3.1: Proposed Diagnostics for the Planar Loading Experiments

Measurement	Diagnostic	Resolution
Single Crystal Laue	Broad Band XRD	
Stress State, Dislocation Density	Monochromatic XRD	10μm, 10ns
Temperature	Neutron Resonance Spectroscopy	10μm, 10ns
Texture and Crystal Structure	Powder -XRD	10μm, 10ns
3D Microstructural Evolution	Coherent X-ray Diffraction Imaging	10μm, 10ns
Early Stage Void/Crack Growth	Small Angle Scattering	0.5μm, 10ns
Late Stage Damage Kinetics	P-Rad	10μm, 10ns
Final Material State	Recovery	

Specimen and Environment:

To address the applied problems associated with the behavior of engineering materials under extreme conditions, examination of specimens that are at least 1mm thick must be possible – although it is important to note that some diagnostics may only work in reflection under these conditions. Additionally, all drive platforms should accommodate experimental temperatures both below and above ambient (77K to 1273K).

3.7.2 Collapsing Cylinder

The deformation path dependence of strength and damage requires exploring a variety stress-states over a broad range of deformation. Certain stress-states, such as uniaxial tension, limit the accessible range of plastic deformation that can be explored due to intervening material failure. Other stress-states, such as uniaxial compression, facilitate large deformations that are useful in characterizing limit-states and understanding strength scaling at large strain. Uniaxial compression of bulk material under typical shock-loading (plate-impact) conditions is useful; however, this geometry strongly couples the mean strain-rate and the ultimate obtainable strain via the Hugniot relation in the strong-shock regime and limited loading flexibility in the weak-shock regime. An alternative geometry that moves away from the shock dominated response is that of a radially collapsing cylinder, with stain-rates of 10^4-10^6 s^{-1}. This configuration allows for a large amount of deformation driven by inertia following the initial loading which could be tailored to incorporate a range of shock loading. By varying the tube radius, thickness, and initial drive conditions a range of off-Hugoniot, post-shock large deformation states could be achieved in a geometry accessible to internal and external probes. The collapsing cylinder experiment may be viewed as a radially symmetric variant of planar loading that facilities impulsive loading followed by a large deformation, lower strain-rate coast phase. To obtain the necessary understanding required to link microstructure to strength and failure, the stress-state, dislocation density, temperature, crystal structure, crystallographic texture, 3D microstructural evolution, early stage void and/or crack growth, and late stage damage kinetics should be measurable with sufficient temporal and spatial resolution. The details of these measurements are discussed in the diagnostics section of the experiment description.

Drives:

A set of loading conditions for this type of experiment can be achieved with a suite of traditional platforms such as direct or indirect high explosive or electro-magnetic drives. To facilitate addressing the fundamental and applied science questions over the pressure–temperature parameter space of interest, peak stresses achievable with these platforms should range from 1 to 500GPa. Pulse durations should range from 1ns to 1μs. Additionally, specimen assemblies or targets should be designed to explore a range of impulsive load and coast paths that include material failure. To provide additional flexibility of impulse loading the capability to design and build graded density intermediate drive tubes should be explored.

Diagnostics:

For the measurements listed above, the following diagnostic probes are suggested to be simultaneously available. It is envisioned that experiments using all or only a subset of these probes will be proposed.

Table 3.2: Proposed Diagnostics for the Collapsing Cylinder Loading Experiments

Measurement	Diagnostic	Resolution
Single Crystal Laue	Broad Band XRD	
Stress State, Dislocation Density	Monochromatic XRD	10μm, 10ns
Temperature	Neutron Resonance Spectroscopy	10μm, 10ns
Texture and Crystal Structure	Powder -XRD	10μm, 10ns
3D Microstructural Evolution	Coherent X-ray Diffraction Imaging	10μm, 10ns
Early Stage Void/Crack Growth	Small Angle Scattering	0.5μm, 10ns
Late Stage Damage Kinetics	P-Rad	10μm, 10ns
Final Material State	Recovery	

Specimen and Environment:
To address the applied problems associated with the behavior of engineering materials under extreme conditions, examination of specimens that are at least 1mm thick must be possible – although it is important to note that some diagnostics may only work in reflection under these conditions. Additionally, all drive platforms should accommodate experimental temperatures both below and above ambient (77K to 1273K).

3.7.3 Expanding Ring/Cylinder

To address fundamental questions regarding the mechanisms and structural evolution during shear localization and ultimate failure in multi-axial loading conditions, we propose expanding ring and cylinder experiments. This class of experiment considers moderate to extreme conditions of the classical expanding ring and cylinder problem to enable both fundamental and applied science questions to be examined. The ring geometry results in a uniaxial stress loading case while the cylinder geometry provides a plane-strain loading case; facilitating a convenient change in loading by simply varying the cylinder length to thickness ratio. To obtain the understanding required to develop a physics-based, predictive capability to model shear localization, failure, and fragmentation the stress-state, dislocation density, temperature, 3D microstructural evolution, and late stage damage kinetics should be measurable with considerable temporal and spatial resolution. The details of these measurements are discussed in the diagnostics section of the experiment description.

Drives:
A set of loading conditions for this type of experiment can be achieved with a suite of platforms such as high explosive, electro-magnetic, or gas-gun (mandrel) drives. To facilitate addressing the fundamental and applied science questions over the pressure–temperature parameter space of interest, peak stresses achievable with these platforms should range up to 10 GPa and drive the material to ballistic velocities. Additionally, specimen assemblies or targets should be designed to explore a range of impulsive load and coast paths.

Diagnostics:
For the measurements listed above, the following diagnostic probes are suggested to be simultaneously available. It is envisioned that experiments using all or only a subset of these probes will be proposed.

Table 3.3: Proposed Diagnostics for the Expanding Ring / Cylinder Loading Experiments

Measurement	Diagnostic	Resolution
Single Crystal Laue	Broad Band XRD	
Stress State, Dislocation Density	Monochromatic XRD	10μm, 10ns
Temperature	Neutron Resonance Spectroscopy	10μm, 10ns
Texture and Crystal Structure	Powder -XRD	10μm, 10ns
3D Microstructural Evolution	Coherent X-ray Diffraction Imaging	10μm, 10ns
Early Stage Void/Crack Growth	Small Angle Scattering	0.5μm, 10ns
Late Stage Damage Kinetics	P-Rad	10μm, 10ns
Surface Temperature / Cracks	High Speed IR and VIS Cameras	

Specimen and Environment:
To address the applied problems associated with the behavior of engineering materials under extreme conditions, examination of specimens that are at least 1mm thick must be possible – although it is important to note that some diagnostics may only work in reflection under these conditions. Additionally, all drive platforms should accommodate experimental temperatures both below and above ambient (77K to 1273K).

3.7.4 Plate Penetration

To address fundamental questions regarding the mechanisms and structural evolution during shear localization and failure, we propose plate penetration experiments. This class of experiment considers moderate to extreme conditions of the classical plate penetration problem to enable both fundamental and applied science questions to be examined. To obtain the understanding required to develop a physics-based, predictive capability to model shear localization, failure, and fragmentation the stress-state, dislocation density, temperature, 3D microstructural evolution, and late stage damage kinetics should be measurable with considerable temporal and spatial resolution. The details of these measurements are discussed in the diagnostics section of the experiment description.

Drives:
Loading conditions for this type of experiment should include: gas/powder gun (single and two-stage) and shape-charge jet to enable penetrator velocities of 100-9000 m/s. To study the incipient stages of the penetration problem, arresting the penetrator is desirable.

Diagnostics:
For the measurements listed above, the following diagnostic probes are suggested to be simultaneously available. It is envisioned that experiments using all or only a subset of these probes will be proposed.

Table 3.4: Proposed Diagnostics for the Plate Penetration Experiments

Measurement	Diagnostic	Resolution
Stress State, Dislocation Density	Monochromatic XRD	10μm, 10ns
Temperature	Neutron Resonance Spectroscopy	10μm, 10ns
3D Microstructural Evolution	Coherent X-ray Diffraction Imaging	10μm, 10ns
Late Stage Damage Kinetics	P-Rad	10μm, 10ns
Final Material State	Recovery	

Specimen and Environment:
To address the applied problems associated with the behavior of engineering materials under extreme conditions, plate thicknesses must be several times that of the projectile diameter. Minimum plate thicknesses of 3mm (10 grains) should be possible. Additionally, all drive platforms should accommodate experimental temperatures both below and above ambient (77K to 1273K). To tune timing and ranges of expected temperatures, the capability to refine such experiments outside the facility proper is desired.

3.8 Exceptional Requirements

- Capability for sample pre-characterization: knowing the 3D static microstructure: texture, chemistry, phase map, chemical banding, dislocation density, residual stress map.
- Drive conditions must be reproducible within 5%.
- Broad Band XRD to allow for single crystal studies and XAFS type measurements.
- Detectors.
- Diagnostic heating of specimens due to probe beams is a concern: shuttering may be necessary.

Section 4

Chemical Kinetics

By Dana Dattelbaum and Larry Fried

4.1 Introduction

Many materials undergo chemical reactions under dynamic compression. For instance, high explosives, polymers, organic liquids, and composites all undergo dramatic transformations under shock loading. The prediction of these responses with a high degree of accuracy and confidence is a core NNSA mission. In addition, there are many opportunities to use extreme conditions to drive chemical reactions and to synthesize entirely new materials with properties unlike those of currently available materials. The study of chemical processes under extreme dynamic conditions has been limited by the lack of diagnostics with a high degree of spatial resolution and the ability to determine the chemical and/or thermodynamic state. High intensity pulsed X-ray sources, such as that potentially offered by MaRIE, promises to overcome many of these limitations, leading to improved understanding and control of dynamic chemical processes.

4.2 Grand Challenges

We have identified three grand challenges in our understanding of chemistry at extreme conditions. These challenges are:

1. How are chemical reactions modified by extreme thermodynamic states and loading paths?
2. How do meso-scale phenomena affect chemical processes?
3. How do hydrodynamics couple to chemical reactions?

Challenge 1: It is known that chemical reactions can either accelerate or decelerate under high pressure loading. Chemical reaction rates are very sensitive to temperature, so the study of chemistry under shock loading is closely related to an understanding of local temperature.

We propose to study chemical reactions by using shock and non-shock loading to achieve pressures of up to 10 Mbar. Laser excitation could also be used to control chemistry during dynamic compression, in order to control chemical synthetic processes. We anticipate that dynamic X-ray diffraction with time resolution between 10 ps and 1 μs will be necessary to interrogate the formation of solid phases during dynamic compression. X-ray spectroscopy will be required to ascertain the form of bonding in materials at extreme conditions.

Work on this problem will help us to determine multi-component phase boundaries and unusual chemical species at extreme thermodynamic conditions. Our fundamental understanding

of detonation, polymer response, and composite response will be substantially improved. There is a strong potential to synthesize entirely new materials, including ultrahard materials, new carbon allotropes, and exotic forms of matter.

Challenge 2: It is well known that the reactions of solids, such as high explosive composites, are strongly affected by the presence of defects, which become localized regions at elevated temperature upon shock loading. The formation of these regions (commonly called hot spots) is not well understood. It is recognized, however, that hot spot formation controls many high explosive safety, initiation, and energy delivery characteristics. Understanding chemistry at the mesoscale opens the possibility of controlling the reactivity of materials through engineered meso-structures that are becoming accessible with additive manufacturing processes.

We intend to study mesoscale phenomena using dynamic X-ray tomography as a primary diagnostic. In addition, a way of determining local temperatures in dynamically compressed materials is very desirable, although there are limitations with currently known techniques. Mesoscale phenomena could be studied *in situ*, using characteristic sample sizes of several cm, or by repeated experiments on very thin samples.

Understanding mesoscale chemistry will enable a new generation of more accurate and predictive high explosive response models. We anticipate a transformation from today's models with narrow regimes of application to a much broader range of applicability, including safety, performance, shock loading, and thermal events. We also anticipate that a new generation of explosive materials could be designed with improved safety characteristics, tenability, and novel

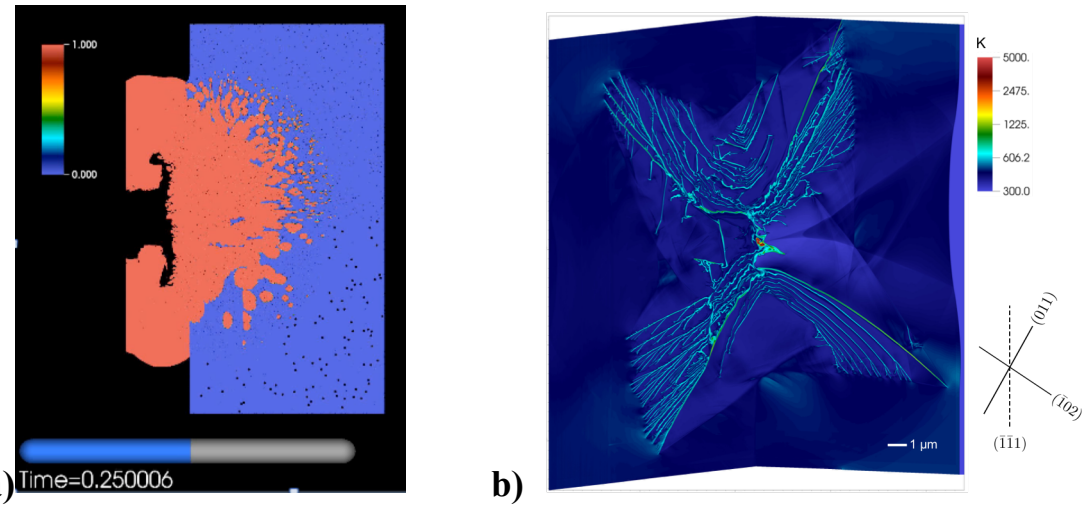

Figure 4.1. Localization of energy at the mesoscale results in non-uniform temperature, strain, density and flow fields which then dictate chemical reaction dynamics and outcomes. A) A calculation of pore effects (2% porosity) on a non-planar reaction front propagation and reactivity in LX-10 impacted by a Kapton flyer. The figure shows the product mass fraction field for LX-10 with 5 micron radius pores impacted at 4.3 km/s. B) Simulation of pore collapse of a single 1 micron pore in crystalline HMX. The figure illustrates the non-uniform temperature fields generated when a strong shock (9.4 GPa) collapses a single small void in the crystalline explosive. Figure from Austin et al. JAP **117**, 185902, 2015

energy delivery capabilities.

Challenge 3: It is known that chemical reactions, which occur on timescales ranging from 100 ps to seconds, strongly couple to hydrodynamics. The coupling between hydrodynamics and

chemistry is responsible for high explosive dead zones and detonation wave failure, the performance of internal combustion engines, and a variety of turbulent mixing processes at extreme conditions. One important open question in this area involves the formation of soots from gaseous combustion or detonation products.

We propose to use small-angle X-ray scattering to resolve the formation of particles and other density changes in real time. We envision a capability where imaging could be combined with small-angle X-ray scattering in order to determine the spatial location of particulate formations. Small angle X-ray scattering could also be combined with X-ray tomography to determine local densities.

The later-stage formation of products of detonation is closely related to high explosive energy delivery, the production of dead zones in high explosives, the performance of combustion engines, and turbulent mixing. Understanding the coupling between hydrodynamics and chemistry will enable greatly improved explosive and polymer models. Mixing processes often limit the performance of inertial confinement fusion systems, so an improved understanding of turbulence and mixing at extreme conditions could enable breakthroughs in inertial confinement fusion design.

4.3 Current Limitations

Existing 3rd and 4th generation light sources, such as the Advanced Photon Source and MEC endstation at the Linac Coherent Light Source (LCLS), are increasingly demonstrating utility toward addressing challenges in chemical dynamics, but in many ways are not ideally suited in their applicability. Particularly within the high pressure/temperature and shock/detonation physics communities, experiments are adapted and designed to conform to the x-ray energies, bunch structures, and facility limitations imposed by existing light sources, rather than having a source that is better matched to the physics issues at hand. Recurrent themes arose across all of the scientific thrusts regarding the limitations of current light sources. These include:

- *Technical limitations in bridging the pertinent length and timescales at which the underpinning physical phenomena are occurring.* For chemical dynamics, the relevant time scales range from ~100 ps to several μs, with a desire to have sub-ns "frames" over long temporal periods during a single experiment.
- *A need for "harder" or higher energy x-rays to increase material penetration depths.* For chemical reactive systems and explosives, the higher energy x-rays are necessary to penetrate thick samples, important due to scaling effects. This limitation is also connected to an ability to study "representative volume elements" and relevant sample sizes at current facilities.
- *High-brilliance, monochromaticity and x-ray energy tunability.* High brilliance and monochromaticity are often traded in current experiments at synchrotron sources to enable sample penetration or improved signal-to-noise, yet complicates diffraction and scattering analysis and interpretation. Furthermore, a high brilliance, monochromatic source will be require for future developments in time-resolved x-ray spectroscopies. These are seen as particularly interesting in chemical dynamics for their ability to distinguish electronic structure of species as they evolve dynamically.
- *Multi-frame and multi-dimensional imaging techniques for real-time tomographic data*. Spatially-resolved density, flow fields (P, ρ, material and wave velocities), mixing phenomena, and local temperatures are key parameters at the mesoscale. Localization phenomena and state dependencies at this length scale dominate the underlying physics, which then dictate the resultant continuum-scale responses. Of seminal importance in chemical dynamics is *temperature*, as the reactions rates have a strong, exponential dependence on temperature, and the

coupling of mesoscale dynamics to local temperature will dominate not only reaction rates, but also equilibria and final reaction product outcomes.

- *Access and throughput*. Facility limitations that currently hinder progress include sample limitations (allowable types and sizes), particularly size limitations on explosive or reactive material charges or hazardous materials, throughput in experimental set-up and execution, which is exacerbated by non-parallelized endstations or hutches at XFELs, and access for defense-related research and development.

Here, the most important of these limitations are the issues of scale due to the state sensitivities of chemical dynamics, and x-ray source characteristics. A harder (~ 40 keV), high brilliance (10^{12} photons) source suited to x-ray diffraction, multi-dimensional imaging, and x-ray based spectroscopies in large, dynamically-driven samples would offer the best matched diagnostic toward addressing grand challenges in chemical dynamics.

4.4 First Experiments

Conceptual experiments were outlined to address physics unknowns and uncertainties identified in the grand challenges, and develop desired requirements of the x-ray source, drive systems, diagnostics, and facility. The experiments were conceived to address both the traditional reactive material challenges with the Department of Energy defense programs, as well as needs of a broader user base within chemical dynamics, including materials synthesis, static high pressure/temperature discovery science, ultrafast laser-based coherent or quantum control of reaction dynamics, and planetary science. Three representative "first experiments" are:

- 3-D investigation of high explosives' initiation at the grain scale
- Chemistry at extreme conditions
- Dynamics of phase segregation and turbulent flows

Experiment #1

The first of these experiments aims to study the coupling of structure and properties at the mesoscale with chemical dynamics and reactivity. This experiment spans several of the grand challenges in understanding how structure couples to dynamics, the effects of hydrodynamics and scale on chemical reactions, and how reactions are modified by thermodynamic state, loading path, deformation and mixing as influenced by structure at the meso- length scale. The experiment is envisioned as a multi-component composite structure with relevant features in the 100s of nm to micron length scales in a mm-size sample that is dynamically loaded and interrogated with a variety of x-ray-based and traditional diagnostics. Examples of the types of samples that are of interest include plastic bonded explosives, granular explosive beds, porous metal oxides or thermites, or other multi-phase composites (rocks/minerals, sand, structural energetics, composite armors, etc.). The sample could be the drive itself, such as the case of an explosive charge, or be driven by a flyer-plate driver (guns, HE, magnetic drive, laser) or direct laser drive with multi-shot rastering capability. Because this experiment probes localization and flow at the mesoscale, and how those then influence chemical pathways and dynamics, multi-dimensional information on local state, turbulence and mixing, and material phases and their evolution are all critical. Diagnostics are envisioned to be multi-probe comprised of tomographic density, temperature (500-5000K+), x-ray diffraction, x-ray scattering, and

spectroscopies, in addition to traditional "bulk" probes such as velocimetry and radiography (x-ray, proton), and optical spectroscopies (absorption, emission, vibrational).

Unique requirements of this experiment that do not exist today include an ability to detonate or study large explosive/reactive material charges (>10g), tomographic density/flow field (with coherent improvements), and x-ray spectroscopies. The experiment would benefit from advancements in scattering and diffraction for morphology characterization and phase identification, respectively.

Experiment #2

One of the most exciting research areas in chemical dynamics is the discovery and synthesis of novel phases of matter formed in extreme conditions at bond energies. Examples include ultrahard materials (such as carbon nitrides), chemical storage materials, novel phases or new allotropes of carbon and carbon compounds, and electride materials. The second experiment aims to access high pressure/temperature phase boundaries, and interrogate and identify unusual chemical species at extreme conditions. Some of the implications of this class of experiments include the potential larger scale synthesis of novel materials, understanding the evolution of chemical species in the context of planetary impacts, and accurate descriptions of species in chemical product mixture equations of state. In these experiments, the drive systems are important for providing both "extreme" and tunable drive conditions. It is anticipated that high compressions reached at multi-Mbar pressures provide access to these unusual states, particularly due to repulsive interactions at high compression and delocalization of electrons. Furthermore, flexible drives, such as non-shock compression drivers will allow for samples to be subjected to high compression/low temperature conditions, which would be advantageous for avoiding ionization at high temperatures. Samples in this experiment would be smaller than in the other two conceptual experiments, and the diagnostic probes would include x-ray diffraction and scattering, and greater emphasis on x-ray spectroscopies (x-ray absorption, Raman, etc.).

Experiment #3

The third experiment focuses on the dynamics of phase segregation and turbulent flows in chemical reactive systems. Key physics and chemistry questions include: 1) how do phases appear/disappear at extreme conditions in reactive systems, 2) how to the phases interact with one another and evolve in time, and 3) how does segregation dictate limiting reaction rates or pathways? Some examples of current topical areas in which

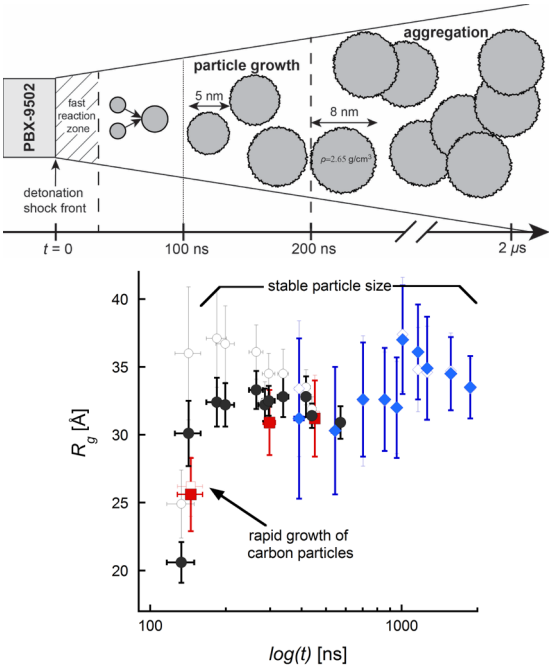

Figure 4.2. The evolution of carbon formation behind a detonation front is an example of dynamic phase segregation in a chemically reacting fluid (figure top). Solid carbon precipitates from a dense fluid product mixture within 100 ns behind the detonation front (Figure bottom), and grows to an average size on the timescales of the chemical reaction zone. Examples shown are for PBX 9502 derived from data taken at the Dynamic Compression Sector, Advanced Photon Source. (LLNL and LANL)

dynamic phase segregation plays an important role in reactive dynamics include carbon precipitation in condensed phase explosives, mixing processes driven by turbulence, the multi-phase chemistry of metal oxides, thermites and intermetallics, and the segregation of species formed in planetary chemistry.

Experimental scale is perhaps most important in this conceptual experiment, as segregation dynamics are determined by thermodynamic state and pathways, which are difficult to attain by miniaturization of experiments to existing platforms. In this experiment, a large experimental sample would be interrogated by a suite of multiple x-ray probes including diffraction and scattering, 3-D imaging, and x-ray spectroscopies with a small region of the larger sample interrogated with exquisite temporal resolution over multiple, many frames. Example samples would include up to 1 kg of explosives, combustion and other reactive material capabilities, including potentially high temperature final product conditions, shock tubes, shock/non-shock-driven reactive samples, and imploding cylindrical samples geometries, particularly for turbulent flows.

Other topics

The conceptual first experiments address several high priority science unknowns identified in the grand challenges. Other topics of interest were also identified by the panel. These included:

- Polymer chemistry: When do polymers break down under loading and how ?
- HE Safety: How can we predict the response of damaged HE ?
- Excited states: Can extreme conditions be combined with coherent laser control ?
- How can we measure conductivity at extreme states ?
- Composites and granular materials: non-uniform reactions occur under shock loading that are not well understood.
- DoD needs: tailorable energetic response, multi-functional materials, structural energetics, resilient armors, agent defeat

4.5 Exceptional Requirements

Exceptional requirements, extending the initial requirements outlined in the MaRIE requirements document for critical decision (CD) 0, were identified to be:

- Bunch repetition rates/bunch structures of 100 ps to 10 ns over 1 microsecond temporal durations
- Multiple (coherent) beams for 3-D tomographic (and possibly spectroscopy) data
- High brilliance, monochromatic x-rays for time-resolved x-ray spectroscopies for chemical species identification
- Focal spot sizes approaching < 1 micron for temporal resolution
- Large explosive charge sizes (up to 1 kg) due to scaling dependencies in reactive flow

Section 5

Mesoscale Structure and Heterogeneous Media

By John Borg and Mukul Kumar

5.1 Introduction

We are surrounded in our physical world by structures that are inherently heterogeneous. These entities, represent challenges where materials and the engineered or biological architectures are constructed such that the collective response is vastly different than simply the sum of the parts. To understand the relevant physical phenomena that determine the *performance* of these structural formations at a fundamental level is a grand challenge because the relevant physical processes can occur over a wide range of time and length scales. This "mesoscale" phenomena (as defined by the DOE-Office of Science) is pervasive and particularly shines through for the case of under-dense materials, such as foams, granular materials, or engineered material lattice structures, where morphology, not composition, is varied to achieve the desired properties. *Having said that, little is understood of the collective response of this class of materials, though extensively studied, as for example, concrete or sand piles, to allow for truly predictive model representations*.

5.2 Background

The behavior of granular materials, as an example of a heterogeneous material system, is complicated in comparison to monolithic counterparts primarily because of the many more degrees of freedom that are accessible to the ensemble. It is therefore not surprising that current continuum based simulations do not capture key details of the granular material response, and consequently lack predictive capability when applied to problems outside the domain of calibration. Similar issues are now being raised as we consider under-dense materials that are somewhat constrained because of the presence of structure, be it of random or regular lattice variety. Examples of these include foams, both of the open and closed cell varieties, and the highly structured lattice materials.

The meso-mechanical response of the broad class of under-dense materials is particularly affected by the early-time behavior during compression, where the collapse through initiation of localization processes in the unique architecture precedes densification, but cannot simply be defined by the materials equation of state (EOS) or constitutive laws. This is a grand challenge in that to achieve predictive power we need to accurately represent the underlying physical processes. The following questions arise:

- When does a heterogeneous system achieve a homogeneous state and vice versa under a strong stress transient, and what is the length and time scale for the transition into a "recognizable" steady shock wave?
- What is the mesoscopic structure of the shock front for heterogeneous media? How does stress propagation through discontinuous media compare between quasi-static conditions, where we can characterize localization in detail, and dynamical conditions, a regime where current models typically sweep over mesoscopic details?
- How does the localization behavior correlate with the correlation length (order parameter) in the mesostructure?

  For example, in structured lattice a two-wave structure is supported up to some number of unit cells, but not in the case of granular systems, as shown in Figure 5.1. On the other hand, a steady wave develops in granular systems once the response is "homogenized" over some length scale, but only a compaction wave has this character in structured lattices, as seen in Figure 5.2. This points to emergent, dissipative phenomena arising from the underlying heterogeneity, which must be understood for us to effectively model the heterogeneous systems.

- What state variables are required to define the thermodynamic state? But these systems also require that we focus not just on mean measurements, but distributions. As an example, consider the stress and strain distributions, shown in Figure 5.3, that are revealed through direct numerical simulations of a particle metal-metal composite.

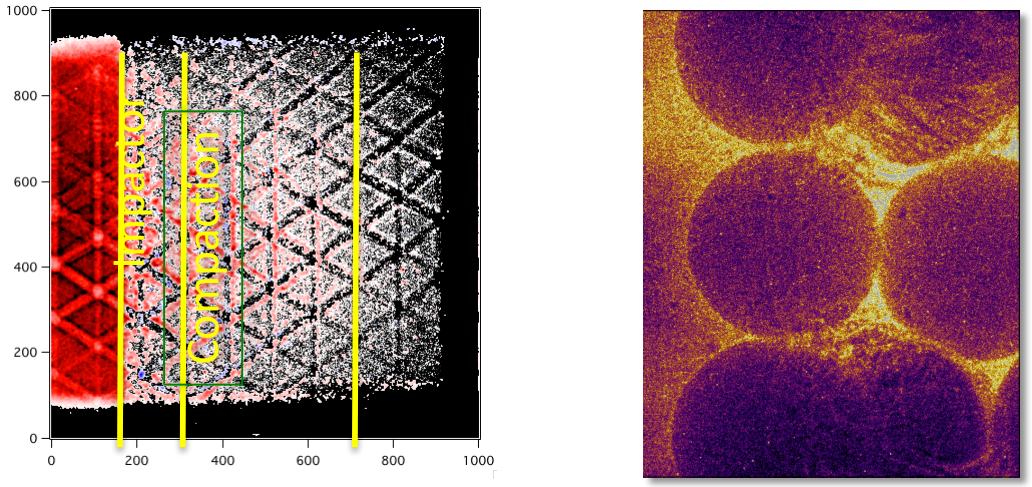

Figure 5.1. Phase contrast images from DCS taken orthogonal to the compression wave, traveling from left to right: on left showing elastic deflection of the underlying 10% dense octet lattice structure, and on right showing a system of granular particles making contact with each other and undergoing local fracture under the action of the traveling compaction wave. (Figures courtesy of Jonathan Lind (left), Minta Akin (right) of LLNL and the IMPULSE collaboration (LANL).)

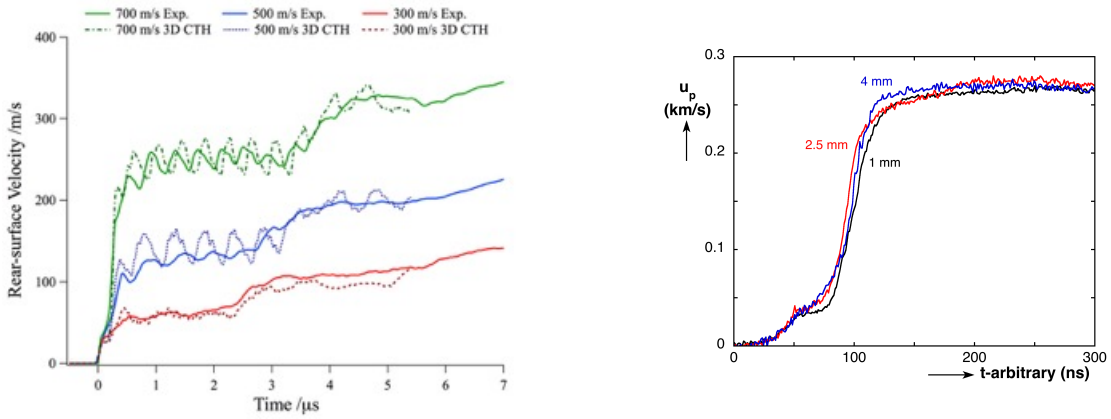

Figure 5.2. Free surface velocities from octet lattice structure (left) and on right from a system of granular particles. The velocity profile shows a lot of structure owing to the breaking of the individual struts, whereas the system with a higher number of degrees of freedom initially develops a steady wave once the response is "homogenized" over some length scale. (Figures courtesy of Daniel Eakins (left) of Imperial College, London and Tracy Vogler (right) of SNL.)

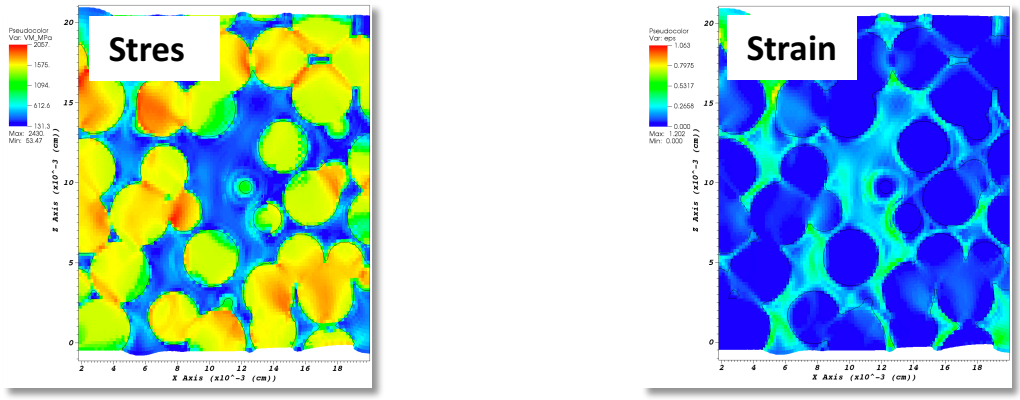

Figure 5.3. Direct numerical simulations of a particle metal-metal composite microstructure with the "hard" phase being the particle and the matrix as the "soft" phase. It is obvious, and intuitive, that the "hard" phase carries most of the stress (left) and the "soft" phase undergoes more plastic strain (right). The challenge is to understand at what length and time dispersive and dissipative mechanisms homogenize into "bulk-like" behavior. (Figures courtesy of Matthew Barham of LLNL.)

5.3 Current Limitations

With regard to experimental measurements, there are barriers to making progress in understanding the dynamic behavior of heterogeneous material systems. Most barriers are fundamentally a result of the small length and time scales associated with the dynamic compression of heterogeneous systems. Obtaining access to these small scales with sufficient

fidelity to resolve a meaningful statistical distributions of the state of the material, which may include deformation, velocity or stress, is challenging. This kind of data would be necessary to build constitutive models of the heterogeneous system's response and move the field of dynamic behavior of heterogeneous systems forward and address the grand challenge questions listed above.

Some mechanical measurements of the bulk system, such as particle velocity, can be made. The response of the system at smaller scales can also be made; however understanding the internal distribution of velocity with enough resolution to understand the statistical nature of the flow remains challenging. For example, line VISAR measurements can tell us something about the variation of the local particle velocity at an interface but do not provide information with regard to the system as a whole. This is especially challenging if one is interested in early time shock formation and breakout as applied to reacting systems. Diagnostic systems that can penetrate the target material cannot provide the sub-nano second temporal resolution and distinguish the material distribution well enough to make local particle velocity measurements.

From the basics of classical steady thermodynamics, one would need to obtain two thermodynamic state variables to fix the thermodynamic state of the bulk system. However, given the nature of heterogeneous systems it is not clear if this is enough to fix the thermodynamic trajectory of all the material within the system. Additional state variables may be needed for example variations in morphology or hysteresis may ultimately affect the behavior of the system. Thus, beyond mechanical measurements, making measurements of the thermodynamic state remains a challenge. For example, knowing the temperature of the material at a point or spatially resolved would be particularly useful in fixing the thermodynamic state of the material. This is especially true when building or validating equations of state. The limitations of measuring temperature are not solely associated with the heterogeneous nature of the material but rather the time scales necessary in the measurement and material accessibility during the experiment. Measuring and spatially resolving temperature variations with pico or even nano-second resolution is challenging. The inherent difficulties of such methods are limited to surface measurements. Given the typical configurations of dynamic experiments limit the optical accessibility of the materials of interest.

5.4 First Experiments

The first campaign seeks to formulate and investigate a canonical problem targeted at better understanding phenomena associated with micro-to-mm heterogeneities, such as those that are present in granular and foam systems. The first materials of interest would be some simple oxide systems such as granular sand (SiO_2) in which one might vary the following characteristics: morphology of the sand grain, particle or pore size distribution, random vs non-random packing. The loading path could be consistent with one-dimensional plain strain experiments such as those derived from plate impact experiments. For example, a planar plate impact loading a system of spherical grains, with a representative volume element that consists of a cube packed with monodisperse 25 μm crystalline particles, to a stress pulse near the crush strength.

The first campaign consists of a series of experiments designed to isolate and vary mechanisms at the mesoscale in order to better understand how these mechanisms at the small scale manifest and emerge at the larger scales. Specifically, these mechanisms include, but are not limited to,

local behavior such as grain contact, interstitial friction, grain morphology, fracture, pore collapse, etc. The objective would be to experimentally isolate a specific mechanism to investigate, such as the effect of mesoscale order in a system or smooth versus rough morphologies in order to isolate given effects. By no means is this a simple task, many careful and laborious experiments are necessary. In addition, careful consideration of the design of experiments is needed such that a reasonable experimental space can be explored.

5.5 Exceptional Requirements

Having the ability to vary the spatial and temporal loading conditions would be useful in exploring phenomenology related to off-Hugoniot behavior. In an ideal situation one would be able to drive the system through any thermodynamic path desired to a given end state over any time frame, thereby connecting static and dynamic loading. Given the physical limitations of experimental methods this may never be realizable. However, it is possible to conceive of and construct experimental techniques to gain access to more of the thermodynamic space than those obtained in uniaxial strain experiments. For example for systems in which the pressure could be increased before launching a shear wave into the system (i.e. pressure-shear experiments), the effects of grain-on-grain friction could be varied in order to assess its effect. Another example would be wave perturbation experiments where the loading could be varied spatially in order to assess the effect of strength on the local flow. Another example is the ability to conduct shockless compression experiments in order to assess the effects of irreversibility so inherent to heterogeneous systems.

With regard to diagnostics, having the ability to make spatially and temporally resolved bulk and local measurements of stress, temperature, local mass/density would be ideal. Coupling this capability with the ability to simultaneously monitor phase information would help formulate a well-posed closed problem.

As one envisions what might be possible and the staggering amount of data that could be generated, a new challenge may be encountered. Such as how quickly one can generate data in a system like that described above? Even if one could obtain all the data requested over any thermodynamic path imagined, one experiment a day would not be sufficient to map a thermodynamic space over some reasonable campaign. How does one correlate the massive amounts of data that would be generated by these types of experiments? Some consideration of the analysis techniques, statistics and/or "data mining" necessary to analyze the data generated could be a serious consideration. This would be tied to new ways of thinking about and analyzing data in order to formulate new theories applicable to heterogeneous systems.

Section 6

Cross-Cutting Themes in the Time Dependent Response of Materials

6.1 Final Thoughts

Our three day international workshop on the Time Dependent Response of Materials brought together over 50 attendees from the DOE and DOD laboratories as well as numerous academic institutions, all with a different perspective regarding the complications associated with time dependencies in dynamic loading environments. This range of perspective is due in part to the breadth of science being discussed, ranging from exothermic reactions in organic molecules, to diffusion limited crystal formation in f-electron materials, to failure in additively manufactured lattice structures. This breadth of topics is generally covered by entirely different academic departments; and yet their time dependent approach towards equilibrium shares a number of cross-cutting themes and therefore pathways to improving our understanding in all the topical areas.

The first crosscutting theme of understanding the formation and evolution of material interfaces can be easily traced through the grand challenges described in:

- Sections 2.2 Understanding the microscopic state of a material with how it pertains to understanding the rate limiting processes for structural phase transitions,
- Section 3.3 Mapping material structure to property relationships by spatially and temporally resolving the evolution of microstructure,
- Section 4.2 Understanding how meso-scale phenomena affect chemical processes,
- Section 5.2 Discovering the mesoscopic structure of the shock front for heterogeneous media and how it affects the continuum response.

While differing broadly in their scientific backgrounds, these disparate fields of dynamic material properties are all looking towards resolving the dynamic mesoscale microstructures. The second crosscutting theme of understanding the dissipative mechanisms associated with the time dependent approach to equilibrium can be seen in the grand challenges of:

- Sections 2.2 Examining the energy flow associated with interfacial motion,
- Section 3.5 Temperature as a diagnostic of material processes during rapid deformation,
- Section 4.2 Understanding how chemical reactions are modified by extreme thermodynamic states,
- Section 5.2 Needing temperature distributions more than average temperature as a required thermodynamic state variable.

Rather than acknowledging our shared complications, these crosscutting themes can be posed as a solution to numerous grand challenge questions within the topical areas that the NNSA considers vital to our core missions. If we could resolve the formation and evolution of material interfaces, we could significantly improve our understanding of how materials change phase, how chemical reactions initiate, how heterogeneous systems flow, and how plasticity correlates with microstructure. If we could track the dissipation of energy as materials undergo non-equilibrium transitions, we could better understand the rate limiting processes associated with rapid phase transitions, and therefore offer a potential avenue to control over the phase transition rates. We could track how chemical reactions are modified by extreme thermodynamic states, such as hot spots, and we could couple thermodynamic distributions to hydrodynamic flows in heterogeneous materials. With an improved understanding of how energy is dissipated, we could better predict how materials flow at extreme strain rates.

Unfortunately, some of the experimental techniques to directly address these cross-cutting themes are many decade long research efforts in themselves, such as spatially and temporally resolved temperature distributions; however, others such as dynamic 3D spatial density distributions are already being obtained at facilities such as the Dynamic Compression Sector at Argonne National Laboratory, Figure 6.1. Unfortunately, we were not able to come to agreement as to whether true spatially resolved measurements are required, or if accurate statistical distributions in density, grain/particle size, and temperature could equivalently progress the field.

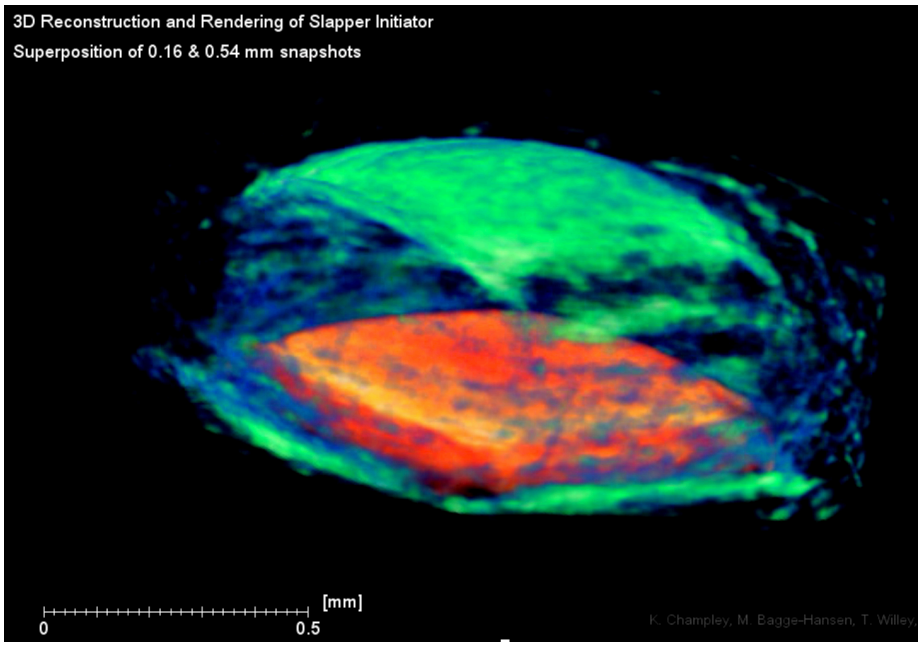

Figure 6.1: 3D tomographic reconstruction of slapper detonator at two points in time (orange-early time, green-late time). Tomographic reconstructions obtained using Livermore Tomography Tools software and multiple angle radiographic views at Dynamic Compression Sector at Argonne National Lab. Figure from T. Willey (LLNL), see Journal of Applied Physics 119, 235901 (2016)

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As it stands, the proposed MaRIE facility could play a tremendous role in advancing our understanding of the time dependent response of materials by enabling us to address these crosscutting themes. MaRIE has a lot of the components our community needs to make significant scientific advances. High-energy coherent x-rays with variable x-ray probe timing would significantly improve our current ability to understand the time dependence of materials. However, we observed a number of technical capability gaps that should be investigated in greater detail.

- Investigating the ability to develop multiple pressure drivers that can span the stress range from 1 GPa to 500 GPa and 1 ns to 1 microsecond loading periods
- Determining the efficacy of 3D dynamic tomography on nanosecond timescales
- Develop a concept for measuring spatially resolved or mass weighted temperature distributions of dynamically loaded materials.

As we look toward NNSA's significant future needs for qualifying materials under extreme static and dynamic environments, advancing these capabilities at MaRIE would make it a transformative facility for our field and the Nation's interests.